<u>AFOSR Nanoelectronics, award no. FA9550-06-1-0270</u>
<u>Title:</u> A multispectral detector based on arrays of carbon nanotubes.
<u>PI:</u> Paola Barbara, Georgetown University
<u>Final report for the period April 1 2006 – June 30 2009</u>

This project focused on studying carbon nanotube quantum dots under terahertz irradiation for potential applications as detectors in the frequency range 0.3-10 THz. The basic physical principle is photon assisted tunneling. A carbon nanotube of finite length has a discrete energy spectrum and can be described as a one-dimensional quantum well. The length of the well is typically the distance between the source and drain electrodes attached to the nanotube. A voltage applied to a nearby gate electrode shifts the Fermi energy in the nanotube by capacitively inducing charges and populating or depleting the energy levels in the nanotube quantum dot. Here we consider the case in which the source and drain electrodes are weakly coupled to the nanotube and a substantial charging energy is necessary to add an electron to the dot. In this case, the current as a function of gate voltage will show sharp peaks corresponding to resonant elastic tunneling of electrons, one at a time, occurring when the Fermi energies of the leads are aligned with an energy level in the dot. Coulomb blockade occurs between the peaks and the voltage interval between adjacent peaks is characterized by the energy level spacing and the charging energy of the dot. When an electromagnetic field is present, transport can also occur via photon assisted tunneling, that is inelastic tunneling with absorption or emission of photons. Photon assisted tunneling generates side peaks in the Coulomb blockade regions: the voltage interval between the side peak and the main peak is determined by the photon energy ($h\nu \cong 4$ meV at 1 THz) and the height of the side peaks is related to the intensity of the electromagnetic field. Carbon nanotubes are ideal quantum dots for photon assisted tunneling in the THz range, because their typical charging energy ($E_C \sim 10$ meV or larger for lengths smaller than a micron) is quite large, allowing detection of side peaks up to a few terahertz at power levels on the order of femtowatts. The lower frequency that can be detected is comparable to the width of the peaks, which depends on the temperature and on characteristic quantum dot properties, such as the coupling between the nanotube and the leads.

Reports of photon-assisted tunneling in quantum dots are scarce [1] and the effect is largely unexplored. This is mainly due to the difficulty to efficiently couple THz radiation to the devices ad the limited availability of powerful THz sources. The goals of our AFOSR supported research were:

1) Design and construction of a variable temperature probe (300K-2K) coupled to a tunable THz source;
2) Fabrication of carbon nanotube quantum dots and quantum dot arrays;
3) Testing quantum dot response to THz radiation as a function of temperature and frequency.

Goals 1) and 2) were accomplished. Goal 3) is still in progress.

Once reliable detection from carbon nanotube quantum dots is obtained, it will be used to detect THz signals with spectral resolution, as well as to fabricate on-chip detectors to test possible operation of carbon nanotube multiple quantum wells as quantum cascade laser THz sources. Below we describe in detail the accomplishments towards goals 1) and 2) and our progress on goal 3).

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Experimental setup

We purchased and tested a BWO source that is tunable in the range 100-180 GHz. The output from this source is coupled to a frequency doubler and a tripler, to achieve tunable frequency in the ranges 200-350 GHz and 600-1000 GHz. The power output at 1000 GHz is about 0.1 mW, two orders of magnitude smaller than a typical laser source. We designed and built a low-temperature probe with optical access through a polished stainless steel waveguide (center tube in Fig. 1.). THz radiation is guided from the source to the waveguide using a 45° reflective mirror, mounted at the top of the probe. The sample needs to be carefully aligned to the focal point of a lens, which is mounted in the cold stage.

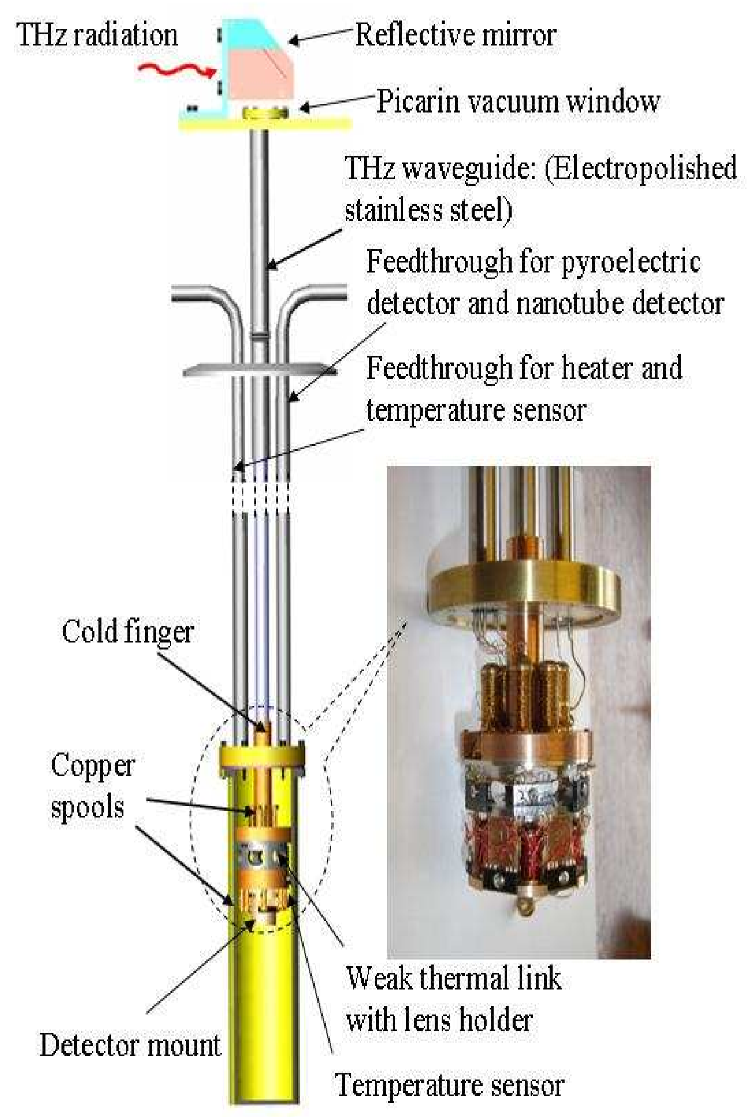

Fig.1. Schematic of our home-made low temperature probe.

Although our probe is a low-cost optical cryogenic system, it requires careful alignment of each sample with respect to the focusing lens mounted in the cold stage. This means that only one chip with a few nanotubes clustered in a small area can be measured for every cooldown. Fig. 2 shows a picture of the actual experimental set-up.

We note that we recently received an AFOSR/DURIP award to purchase a cryostat with optical windows that can operate from room temperature to 3 K without the use of liquid helium. This cryostat will certainly speed up sample testing, because optical windows provide easier alignment between samples and THz source, and will also eliminate expenses associated with liquid helium consumption.

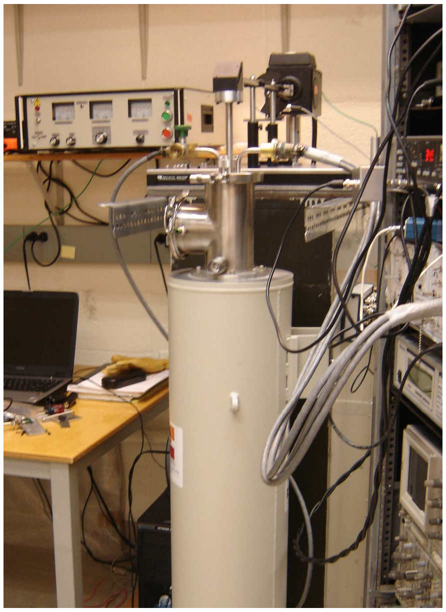

Fig.2. Image of the actual set-up, including the low-temperature probe (see Fig. 5) and the THz source.

Sample fabrication and characterization

A field emission scanning electron microscope with e-beam lithography was acquired and installed at Georgetown University in the Summer 2006, following a NSF/MRI award (PI: P. Barbara). We have developed device patterning and alignment techniques and started fabricating carbon nanotube transistors ranging from 100 nm to 1 μm.

The nanotubes are grown by chemical vapor deposition from patterned catalyst islands and source and drain electrodes are patterned using both e-beam lithography, for submicron features, and shadow masks for larger contact pads. The source and drain electrodes are formed by sputtering thin films (50 nm) of palladium or niobium. In our first quantum dot design, the doped silicon substrate, capped with 500 nm of silicon dioxide, is used as a gate electrode.

We characterized nanotube junctions from room temperature to 2K. The energy levels in the quantum dots were determined by measuring differential conductance of the devices as a function of gate and source-drain voltage, as shown in Figs. 2 and 3, and all the nanotube junctions showing quantized energy levels have been tested under exposure to THz radiation. Our THz source can be tuned in a wide frequency range (100-180 GHz, 200-350 GHz and 600-1000 GHz) and the exposure to THz radiation is controlled by an optical chopper. The signal from the chopper is fed as a reference to the lock-in amplifier, which is used for the differential conductance measurements of the junctions. Although our samples are qualitatively very similar to the samples measured in ref. [1] and show good quantum dot behavior, all the samples we tested so far did not show a measurable response to THz radiation. This can be explained considering that our THz source is two orders of magnitude weaker than the laser source used in the detection experiment in ref. [1]. Moreover, even though the source used in ref. [1] was quite powerful, only few samples showed a response [2], indicating that the reliability of the response is still an unsolved issue.

Progress on improving coupling to THz radiation

This past year we focused on optimizing the coupling between the THz field and the CNT quantum dots according to the following plan:

- Change device design to include on-chip antennas to improve coupling with THz radiation.
- Use more powerful laser sources to better test coupling with different antenna designs.
- Use top electrodes as local gates to create tunable barriers in the dot and obtain arrays of tunable quantum dots with individual side gates, to explore photon assisted tunneling as a function of quantum dot parameters.
- Measure response as a function of temperature and frequency.

We worked on the first three points and fabricated new samples that are ready to be tested at low temperature.

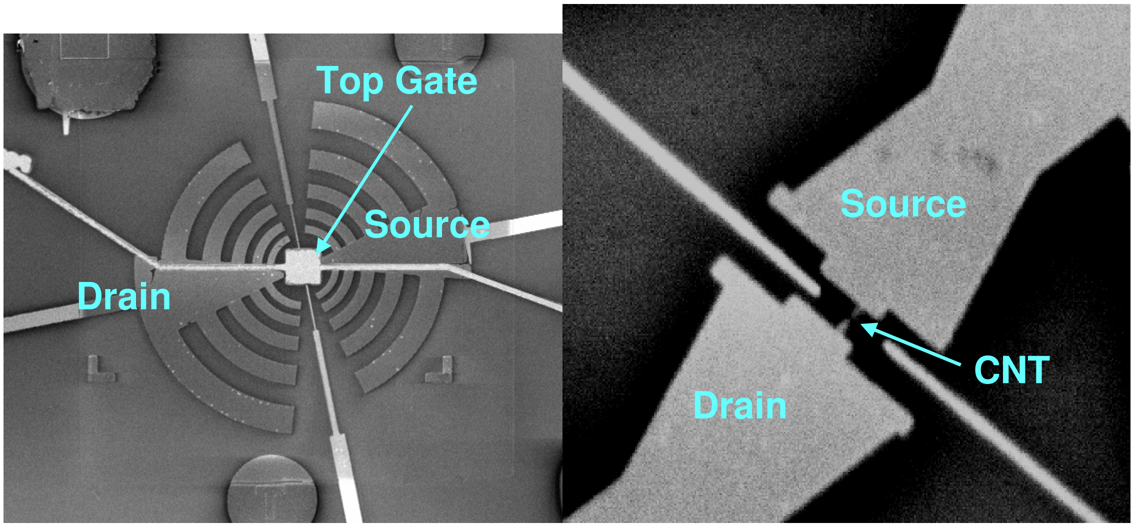

Fig. 3: Left: Log periodic antenna design to operate in the frequency range 700 GHz to 2.5 THz. The antenna was patterned by e-beam lithography and liftoff of 100 nm thick sputtered Gold. Right: Enlarged area showing the source and drain electrodes connected to the carbon nanotube before the deposition of the top electrode. The distance between the source and drain electrodes is about 400 nm.

In addition to high reflection losses, due to the mismatch between the detector impedance and the free space impedance, the dimension of the effective detection region (which is comparable to the size of the quantum dots, about ~ 300 nm) is much smaller than the radiation wavelength (300 microns at 1 THz in free space), as well as the region in which the THz power can be focused (~ 5 mm^2). **On-chip antennas** therefore are necessary to couple the low-power THz radiation to carbon nanotube devices. Antennas will significantly increase the effective detection area and will reduce impedance mismatch. Since we plan to test the device response as a function of frequency, we select antenna designs that work in a wide frequency range. Planar self-complementary antennas [3] are good candidates because their impedance is frequency independent and close to the impedance of free space (within about a factor of two). One example is shown in Fig. 3, where we patterned electrodes as log-periodic antennas [4], designed to operate in the frequency range 700 GHz to 2.5 THz. For these samples the substrate is intrinsic silicon and the gate electrodes will be patterned either on the side of the nanotube or on the top, after growing a SiO_2 layer or Al_2O_3 layer on top of the device.

We plan to test these antennas coupled to the nanotubes with a **more powerful laser source.** We recently started a collaboration with Prof. Dennis Drew at the University of Maryland. The laser lines available in his laboratory and their corresponding power output are listed in the table below.

CO_2 pumped Laser Lines Available at UMD

Frequency (THz)	FIR Gas	FIR Output ~ (mW)
7.2	CD_3OH	8
6.9	CD_3OH	8
5.3	CH_3OD	21
4.3	CH_3OH	12
3.1	CH_3OH	30
2.5	CH_3OH	50
2.3	CH_3OH	10
1.5	CH_3NH_3	5

Lower frequencies can be obtained by providing additional gases. For example, a 1.28 THz line can be obtained by purchasing CH_2F_2.

The laser will be coupled to the sample (which is at liquid helium temperature) either by using the low temperature probe built in our laboratory or a cryostat with optical windows available in Prof. Drew's laboratory. We already adapted this cryostat to mount carbon nanotube quantum dots on the sample stage. A Winston cone is mounted behind the optical window to focus THz radiation on the on-chip antenna (see Fig. 4).

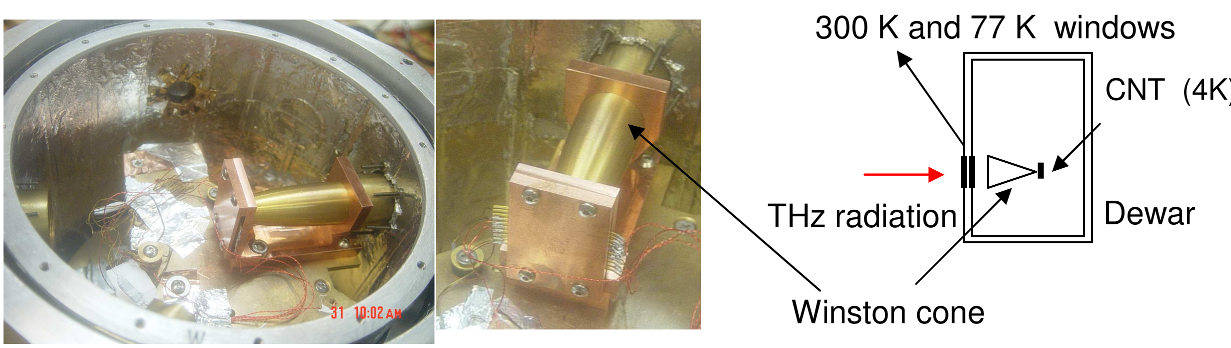

Fig. 4: Mounting of the CNT device for illumination with THz radiation. A Winston cone as used to concentrate the beam on device area. This configuration will also reduce the possibility of optical misalignment considerably. Each polyethylene window has ~ 85 % transmittance below 20 THz.

As we mentioned earlier, we also changed the design of the quantum dots connected to the antennas. We previously tested quantum dots with the doped silicon substrate used as a backgate and the length of the quantum dot defined by the distance between source and drain electrodes. Although these devices have a simpler fabrication procedure (no gate electrode needs to be patterned) and show good quantum dot behavior, **new quantum dot designs need to be developed**. The use of a doped silicon substrate instead of intrinsic silicon will substantially change the performance of the on-chip antenna. We started using substrates made of intrinsic silicon. In this case, we pattern small top gate

that cover just the quantum dot area or small side gates (see Fig. 3). The top gates are patterned after depositing an insulating layer on the device. We tested several options for the insulating layer, including PECVD deposition of SiO_2 in our laboratory or ALD of Al_2O_3 at the Nanocenter at the University of Maryland. Several tests showed that the ALD process is best suitable for fabrication of carbon nanotube quantum dots. Testing of the new samples is ongoing.

References

[1] Y. Kawano, S. Toyokawa, T. Uchida and K. Ishibashi, *THz photon assisted tunneling in carbon-nanotube quantum dots*, Journal of Applied Physics **103**, 034307-034310 (2008).

[2] Dr. Koji Ishibashi, private communication.

[3] Y. Mushiake, *Self-complementary antennas,* IEEE Antennas and Propagation Magazine, **34**(6), 23–29, 1992.

[4] R. H. DuHamel & D. E. Dummel, *Broadband Logarithmically periodic antenna structures*, IRE International Convention Record, vol. 5, pp. 119-128, (1957).